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# ANALYSIS OF DRY STORAGE TEMPERATURE LIMITS FOR ZIRCALOY-CLAD SPENT NUCLEAR FUEL

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## ABSTRACT

Safe interim dry storage of spent nuclear fuel (SNF) must be maintained for a minimum of twenty years according to the Code of Federal Regulations. The most important variable that must be regulated by dry storage licensees in order to meet current safety standards is the temperature of the SNF. The two currently accepted models to define the maximum allowable initial storage temperature for SNF are based on the diffusion controlled cavity growth (DCCG) failure mechanism proposed by Raj and Ashby. These models may not give conservative temperature limits. Some have suggested using a strain-based failure model to predict the maximum allowable temperatures, but we have shown that this is not applicable to the SNF as long as DCCG is the assumed failure mechanism. Although the two accepted models are based on the same fundamental failure theory (DCCG), the researchers who developed the models made different assumptions, including selection of some of the most critical variables in the DCCG failure equation. These inconsistencies are discussed together with recommended modifications to the failure models based on more recent data.

## INTRODUCTION

Interim dry storage of spent nuclear fuel (SNF) rods is of critical concern because of the shortage of wet storage capacity and delays in the availability of a permanent disposal repository. Safe dry storage must be maintained for a minimum of twenty years [1]. The NRC has approved two models [2,3] to determine the maximum initial temperature limit for nuclear fuel rods in dry storage that supposedly meet all safety criteria [1] and yield consistent temperature limits. Though these two models are based on the same fundamental failure theory, different assumptions have been made including the choice of values for material constants in the failure equation. This paper will discuss these inconsistencies as well as some of the shortcomings of the current models and suggest some modifications.

## CURRENT MODELS

Currently, the maximum allowable initial temperatures for interim dry storage of SNF rods are determined using either the equations developed by Lawrence Livermore National Laboratory (LLNL) [2] or temperature limit curves developed by Pacific Northwest National Laboratory (PNNL) [3]. Both the PNNL and the LLNL models predict that cavitation failure under dry storage conditions may occur by diffusion controlled cavity growth (DCCG). Many investigators (e.g., [4]) have suggested that under conditions similar to dry storage, the failure mechanism may not be exclusively DCCG, but rather a coupled diffusion and power law creep mechanism. Without large far field strains, however, (as is absent in the case of zircaloy fuel rods [5,6]), cavity growth does not seem possible by power law creep. Thus, we tentatively concluded that a diffusion controlled cavity growth model is the most reasonable.

LLNL uses the DCCG analysis by Raj and Ashby [7] to predict the time to failure of zircaloy under dry storage conditions based on a 'limited damage' approach. The equations can be used to calculate initial temperature limits for SNF for specified temperatures and stresses. PNNL predicted temperature limits using a fracture map to account for various fracture mechanisms predicted to be active over a relevant range of stresses and temperatures. For dry storage temperatures, the fracture map indicates that either DCCG (~0-110 MPa) or power law creep (~110-160 MPa) controls failure. The most relevant stresses for dry storage are less than 110 MPa, so both models predict DCCG controls failure for dry storage of SNF.

Some have instead suggested (e.g., [5,8]) using a creep-strain limit approach where creep-strain is limited to 1% which is prior to tertiary creep. A creep-strain limit was chosen based on the proposition that fracture only occurs during tertiary creep. DCCG, however, is not strain dependent. It has been shown (e.g. [9, 10]) that cavities can nucleate and grow in various metals through *all* stages of creep.

### **Inconsistency between LLNL and PNNL models**

The two models predict nearly consistent temperature limits for stresses most relevant to dry storage (from about 40-100 MPa). However, it turns out that the near coincidence of the model predictions results from a fortuitous combination of differing assumptions by PNNL and LLNL. First, PNNL (but not LLNL) used a 'recovery factor' in all failure equations. This factor is used to account for recovery of some of the reduction in ductility of zirconium alloys from irradiation damage. Because the DCCG fracture model is *not* a function of strain, a reduced ductility would not directly affect the fracture time. It is not clear, therefore, whether a "recovery factor" should be used with DCCG. Additionally, LLNL chose a value of 0.15 (15%) "area fraction of decohesion" to correspond to failure based on estimated post-dry-storage handling of the fuel rods [11]. The PNNL model, however, incorporates the original form of the Raj and Ashby [7] failure equation which inherently assumes that failure occurs at an area fraction of decohesion of 0.50 (50%). Finally, PNNL and LLNL chose different values for the material constants used in the DCCG failure equation. These differences include atomic volume,  $\Omega$ , grain boundary thickness,  $\delta$ , cavity spacing,  $\lambda$ , grain boundary diffusion coefficient,  $D_o^{gb}$  and  $Q_{gb}$ , as well as other unspecified constants.

### **Temperature sensitivity of current models**

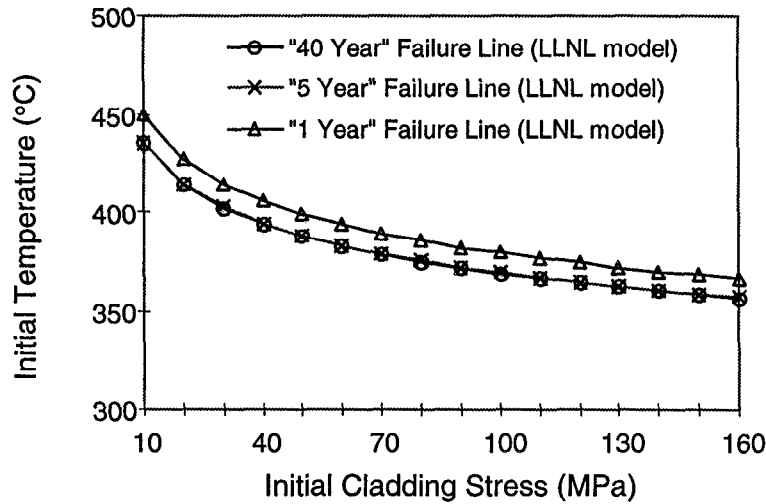
Both models are very sensitive to changes in the temperature decay of the SNF. A calculated initial temperature thought to cause failure in 40 years essentially predicts failure in 5 years for the *same* initial temperature when using Equation 1 below, which PNNL suggests is reasonable. The temperature profile used by PNNL when comparing to the LLNL model is

$$T(K) = C \times [t(\text{months})]^{-0.282} \quad (1)$$

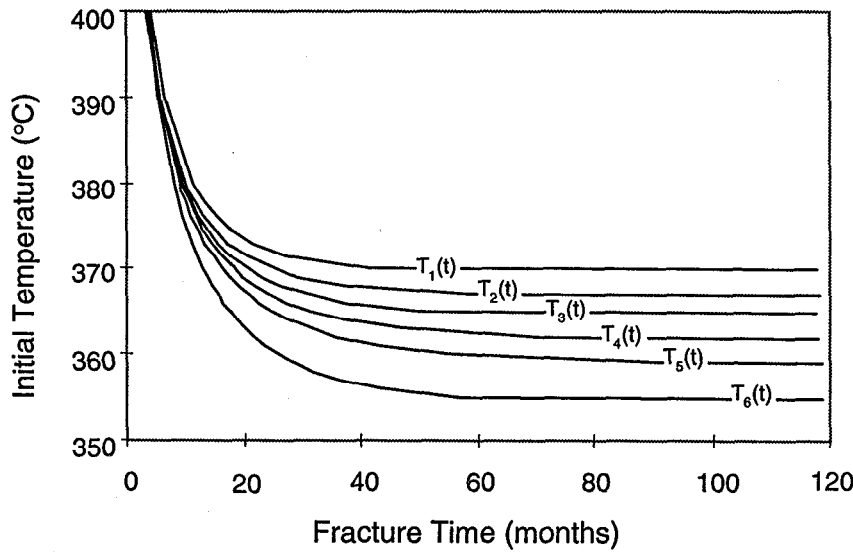
where C can be adjusted to modify the initial temperature of the SNF. This calculates the temperature of the SNF as a function of time after removal from reactor.

The DCCG failure equation with this assumed temperature profile predicts that nearly all of the damage will occur in the first 5 years. If a power type temperature profile such as equation 1 is the best choice, then both (PNNL and LLNL) models predict that if failures are going to occur, they will occur within the first 5 years in dry storage. Again, a failure according to the LLNL criteria implies that the material has experienced a reduced cross sectional area, not necessarily a rupture. Thus, care must be exercised when evaluating the DCCG failure equation

for long storage times. This limitation is why the predicted maximum allowable temperatures can be misleading, as illustrated in Figure 1.



**Figure 1.** Comparison of 40, 5 and 1 year failure lines predicted using the LLNL model.



$$\begin{aligned}
 T_1(t) &= C1 \times 10^3 t^{-0.282} \text{ (K)}, \quad T_2(t) = C2 \times 10^3 t^{-0.26} \text{ (K)}, \quad T_3(t) = C3 \times 10^3 t^{-0.24} \text{ (K)} \\
 T_4(t) &= C4 \times 10^3 t^{-0.22} \text{ (K)}, \quad T_5(t) = C5 \times 10^3 t^{-0.20} \text{ (K)} \\
 T_6(t) &= T_1(60) + [T_1(180) - T_1(60)]/2 - [T_1(180) - T_1(60)]/120 t \text{ (K)} \quad \begin{cases} t < 180 \\ t \geq 180 \end{cases} \\
 &= T_1(180) \text{ (K)}
 \end{aligned}$$

**Figure 2.** Fracture time as a function of initial storage temperature for various temperature decay functions for 5-year-old SNF with an initial cladding hoop stress of 100 MPa.

When using a temperature profile similar to Equation 1, small changes in the initial temperature result in large differences in the predicted failure time. Alternatively, large changes in the specified failure times result in insignificant changes in the maximum allowable initial temperature (e.g., failure times ranging from 5 years to infinity). Therefore an applicant who determines a maximum temperature for a predicted failure of 300 years has, at the same time,

predicted failure after 5 years (which is *less* than the licensed period of the storage facility). Figure 2 illustrates how more conservative temperature decay profiles would affect the predicted failure time based on the LLNL model. The sensitivity of the model is reduced with each successively less concave temperature decay profile (1-5). The time over which 99% of the damage occurs similarly increases (from 5 years with profile 1 to 9 years with profile 5). This is true because each successively less concave profile indicates more time at higher temperatures resulting in higher diffusion coefficients and stresses, causing more cavitation of the material. The linear profile is the most conservative. It predicts that more damage occurs (shorter fracture time) than any of the power decay profiles. It also decreases the sensitivity of the failure time to the initial temperature of the SNF compared to the temperature decay profile suggested by PNNL.

Although measured profiles seem to obey power-like temperature decay, the uncertainty involved in developing the temperature profile (see reference 12 discussion) and the fact that the predicted failure time is so sensitive to this profile suggest using a conservative profile. We therefore recommend using an accurate temperature profile measured under the proposed storage configuration or, if unavailable, using a conservative linear decay profile.

### **Critical material parameters**

#### ***Grain boundary diffusion coefficient***

Self-diffusion coefficients reported in the literature for zirconium (zircaloy not available) vary by more than four orders of magnitude (see [12] for references). Choosing a grain boundary diffusion coefficient based on these data, as PNNL and LLNL have done, is not straightforward. PNNL suggests an activation energy,  $Q_{gb}$ , of 175 kJ/mole and a value for  $D_o^{gb}$  of  $3.89 \times 10^{-6}$  m<sup>2</sup>/sec (basis not reported). LLNL suggests using the activation energy,  $Q_{gb}$ , of 131 kJ/mole and  $D_o^{gb}$  of  $5.9 \times 10^{-6}$  m<sup>2</sup>/sec reported by Garde et al. [13]. This yields a diffusion coefficient that is 3 to 5 orders of magnitude faster than PNNL's suggested  $D_{gb}$  at 400°C and 200°C, respectively. At the time these models were developed, no grain boundary diffusion studies had been reported for zirconium. The values chosen by LLNL and, presumably, by PNNL were based on the approximate relationship between the self-diffusion and grain boundary diffusion activation energies  $Q_{gb} \approx 0.6Q_{sd}$  [14]. This is one serious limitation with these models as the more than four order of magnitude variation in reported values for the diffusion coefficient translates linearly to the predicted failure time. Since the time that these models were developed, however, grain boundary diffusion data have been reported. Vierregge and Herzig [15] measured the grain boundary diffusion coefficient of zirconium. This study gives  $D_{gb}$  as

$$D_{gb} = 4.2_{-2.5}^{+5.9} \times 10^{-13} \exp\left(\frac{-167 \pm 7 \text{ kJ/mole}}{RT}\right) \delta \frac{\text{m}^2}{\text{s}} \quad (2)$$

where  $\delta$  is the grain boundary width. The predictions of Equation 2 are close to that suggested by LLNL, however the error indicated in Equation 2 translates to a variation of more than 100°C in the maximum initial temperature when using Equation 1 for the temperature profile. Furthermore, it is not clear what effect irradiation may have on the grain boundary diffusion coefficient of zirconium alloys. Irradiation damage results in a higher concentration of vacancy type dislocation loops near grain boundaries [16,17] and also causes dispersion of Fe which could substantially increase the diffusion rate (see reference 12 discussion). The effect that

alloying zirconium on grain boundary diffusion under conditions relevant to dry storage has not been established, however zircalloys are richer in iron than zirconium and iron has been shown to dramatically increase the self-diffusion coefficient of zirconium.

### ***Cavity spacing***

PNNL assumes a constant cavity spacing of 2.6  $\mu\text{m}$  (basis not reported) and LLNL assumes a constant spacing of 10  $\mu\text{m}$  based on [9]. These two suggested values may not be conservative. There is evidence that irradiation may lead to a cavity spacing of about 0.1-0.2  $\mu\text{m}$  [17]. It may be reasonable, therefore, to assume a 0.1-0.2  $\mu\text{m}$  cavity spacing.

## **ROD SURVEILLANCE TESTING**

It has been suggested [5,18] that SNF stored under current dry storage conditions appears safe over extended periods of time based on rod surveillance testing. Post-storage integrity (a handling requirement after dry storage), however, has not (to our knowledge) been evaluated after long term storage. Furthermore, rod surveillance does not allow quantification of stresses and temperatures. Also, the microstructure of these fuel rods was not examined after dry storage. It is not known whether the condition of the fuel rods is still safe according to the current safety standards. The established 15% area fraction of decohesion limit may have been surpassed *without* rupture occurring in dry storage, but the rods may be unsuitable for post dry storage handling [11].

## **CONCLUSIONS**

Although based on the same fundamental fracture theory (DCCG), the two currently accepted models (by PNNL and LLNL) include many inconsistent assumptions, including the assumed failure condition and choice of values for constants in the DCCG failure equation. Many of the inconsistent assumptions arise because both models include the variables that have not been accurately measured for irradiated zircaloy (e.g., grain boundary diffusion coefficient and cavity spacing). Using the limited data available for zirconium is tenuous, as the effects of alloying on the grain boundary diffusion coefficient of zirconium and the effects of irradiation on the diffusion coefficient and the cavity spacing have not been established. These factors combined with extreme sensitivity to temperature leave the current models with marginal value. It should not be assumed that these models yield conservative temperature limits until experimental data for cavity spacing, grain boundary diffusion and post-dry storage integrity for irradiated zircaloy is obtained. A strain-based model is not applicable as long as DCCG is the assumed fracture mechanism. Long term creep testing under dry storage conditions to determine the operating fracture mechanism and grain boundary diffusion measurements on irradiated zircaloy should be started immediately.

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